The recent observation of superconductivity with critical temperatures ($T_c$) up to 55 K in the pnictide $\text{RFeAsO}_{1-x}F_x$, where $R$ is a lanthanide, marks the first discovery of a non-copper-oxide-based layered high-$T_c$ superconductor$^{1-3}$. It has raised the suspicion that these new materials share a similar pairing mechanism to the cuprate superconductors, as both families exhibit superconductivity following charge doping of a magnetic parent material. In this context, it is important to follow the evolution of the microscopic magnetic properties of the pnictides with doping and hence to determine whether magnetic correlations coexist with superconductivity. Here, we present a muon spin rotation study on SmFeAsO$_{1-x}$F$_x$, with $x = 0-0.30$ that shows that, as in the cuprates, static magnetism persists well into the superconducting regime. This analogy is quite surprising as the parent compounds of the two families have rather different magnetic ground states: itinerant spin density wave for the pnictides contrasted with the Mott–Hubbard insulator in the cuprates. Our findings therefore suggest that the proximity to magnetic order and associated soft magnetic fluctuations, rather than strong electronic correlations in the vicinity of a Mott–Hubbard transition, may be the key ingredients of high-$T_c$ superconductors.

Similar to the cuprates, the pnictide high-critical-temperature ($T_c$) superconductors (HTSCs) have a layered structure comprising alternating FeAs and LaO sheets, with the Fe arranged on a square lattice$^1$. Theoretical calculations predict a quasi-two-dimensional electronic structure, with LaO layers that mainly act as blocking layers and metallic FeAs layers that are responsible for superconductivity$^{4-6}$, although these are multiband superconductors with up to five FeAs-related bands crossing the Fermi level$^7$. Like the copper-oxide HTSCs, the superconducting state in the pnictides emerges on charge doping a magnetic parent compound$^{8-10}$, with indications that the maximal $T_c$ occurs just as magnetism disappears$^{11-13}$. The last point may well be of great significance, as the parent compounds in the two families are very different. For the pnictides, there are strong indications that they are itinerant systems with magnetism arising from a nesting-induced spin density wave (SDW). This is in contrast to the cuprates, where it is well established that the mother compounds are ‘charge transfer insulators’, where strongly repulsive electronic correlations yield an insulating and antiferromagnetic ground state despite a half-filled conduction band. It is therefore of great importance to obtain further insight into the differences and similarities of the pnictide and cuprate HTSCs. A particularly important question is how magnetism and superconductivity evolve on electron doping. In this context, muon spin rotation ($\mu$SR) is an ideal technique as it provides microscopic information corresponding to the bulk of a sample and there is a substantial body of $\mu$SR data that has been collected on the copper-oxide HTSCs for comparison$^{14-16}$.

Here, we present our zero-field $\mu$SR (ZF-$\mu$SR) measurements, which detail how magnetism evolves with F doping from the SDW state below $T_{\text{mag}} \approx 135$ K in the parent compound at $x = 0$ towards the superconducting state at $x \geq 0.1$. In particular, we show that static magnetic correlations originating from the FeAs layers survive to a surprisingly high doping level well into the superconducting regime. Figure 1a–e shows representative ZF-$\mu$SR spectra, which show the time dependence of the muon spin polarization, $P(t)/P(0)$, at temperatures below and above $T_{\text{mag}}$ in the order of increasing F content for $0 \leq x \leq 0.13$. The solid lines show fits to the experimental data with the function:

$$P(t) = f_1 \left( \frac{2}{3} G_{\text{osc}} + \frac{1}{3} \right) \exp(-\lambda_1 t^\beta_1) + f_2 \exp(-\lambda_2 t^\beta_2) + f_3$$

The first term with a fraction $f_1$ represents the static or quasi-static magnetic signal, the second term with a fraction $f_2$ the dynamic magnetic signal and the third term a fixed background component due to about 10% of the muons that miss the sample and stop in the sample holder or the cryostat walls. For $x = 0$, the oscillatory part of the magnetic signal takes the form $G_{\text{osc}} = \cos(\omega_0 t) \exp(-\sigma^2 t^2)$, whereas $G_{\text{osc}} = f_0(\omega_0 t) \exp(-\sigma^2 t^2)$ for $x > 0$. Here, $\omega_0$ is a characteristic precession frequency, $\sigma$ is a Gaussian relaxation rate reflecting the frequency width and $f_0$ is a zeroth-order Bessel function. The parameters $\lambda_{1,2}$ and $\beta_{1,2}$ describe the generalized relaxation of the first two components due to magnetic fluctuations. The Bessel function used for $x > 0$ reflects the intrinsic distribution

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Figure 1 | Temperature and doping dependencies of the ZF-\(\mu\)SR spectra of SmFeAsO_{1-x}F\(_x\). a–e, Time-dependent spectra of the muon-spin-polarization, \(P(t)/P(0)\), at representative temperatures below and above the magnetic ordering temperature, \(T_{\text{mag}}\), for polycrystalline SmFeAsO_{1-x}F\(_x\) samples with \(x = 0\) and \(T_{\text{mag}} \approx 135\) K (a), \(x = 0.05\) and \(T_{\text{mag}} \approx 80\) K (b), \(x = 0.1\), \(T_{\text{mag}} \approx 60\) K and \(T_c = 10(7)\) K (c), \(x = 0.12\), \(T_{\text{mag}} \approx 35\) K and \(T_c = 17(8)\) K (d) and \(x = 0.13\), \(T_{\text{mag}} \approx 30\) K and \(T_c = 25(8)\) K (e). Symbols show experimental data and solid lines fits with equation (1) for which the obtained parameters are shown in Fig. 2a–c.

Figure 2 | Evolution of the magnetic signal of the \(\mu\)SR measurements on SmFeAsO\(_{1-x}F\(_x\)\) as a function of doping and temperature. a, Doping dependence of the local magnetic field, \(B_\mu\), and the relaxation rate, \(\lambda_{\text{ZF}}\), as obtained from fitting the ZF-\(\mu\)SR data. The red shaded area marks the superconducting region. b, Doping dependence of the amplitudes \(f_1\) and \(f_2\) in the ZF-\(\mu\)SR spectra together with the superconducting transition temperatures, \(T_c\), as obtained from resistivity and magnetization measurements. Inset: Magnetic volume fraction \(f_1 + f_2\) obtained from the low-temperature weak transverse-field \(\mu\)SR measurements (using equation (2)). c, Temperature dependence of the local field at the muon site, \(B_\mu\), as deduced from the precession frequency, \(\nu_\mu\), in the ZF-\(\mu\)SR spectra with \(\nu_\mu = (\gamma_\mu B_\mu / 2\pi)\) and \(\gamma_\mu = 2\pi\times 135.3\) MHz T\(^{-1}\) being the gyromagnetic ratio of the muon. Arrows indicate our estimates of the magnetic transition temperature, \(T_{\text{mag}}\).
of internal fields associated with an incommensurate SDW (ref. 17). The temperature and doping dependencies of the fitted parameters are summarized in Fig. 2a–c. They establish that bulk and static or quasi-static magnetism persists to a doping level of about $x = 0.13$ and thus survives well into the superconducting regime.

In the first place, this raises the question of whether the magnetism originates from electronic moments within the FeAs layers or is rather due to the ordering of the Sm moments. Our ZF-$\mu$SR data establish that all samples for $x \leq 0.13$ exhibit fairly high magnetic transition temperatures of $T_{\text{mag}} \geq 30$ K. This is a clear indication that the magnetic moments originate from the FeAs layers and are not associated with the Sm ions, which order at much lower temperature\(^\text{16}\). The absence of a significant contribution of the Sm moments to the magnetic order at $T \geq 10$ K is also confirmed by our ZF-$\mu$SR data on the parent material SmFeAsO\(_3\), of which the precession frequency of 23.6 MHz (see Fig. 2c) is very similar to the frequency observed in LaOFeAs (ref. 10). The static magnetism above 5 K is therefore due to magnetic moments that originate from the FeAs layers. As outlined in Supplementary Information, the ordering of the Sm moments below 5 K is evident in the ZF-$\mu$SR data, as well as in our specific heat data for the $x = 0.10–0.13$ samples.

To obtain a quantitative analysis of the magnetic volume fractions, we have carried out further weak transverse-field $\mu$SR measurements. In a weak transverse field, the magnetic fraction is not affected by the applied field $B$ and only the non-magnetic terms in equation (1) precess in response to the field, giving the following polarization function:

$$ P(t) = f_1 \left( \frac{2}{3} G_{\text{loc}} + \frac{1}{3} \right) \exp(-\lambda_1 t) + f_2 \exp(-\lambda_2 t) t^2 \\
+ f_0 \cos(\gamma_0 Bt) $$  \hspace{1cm} (2)

Any non-magnetic fraction gives rise here to a weakly damped oscillatory signal with an amplitude that can be readily determined. For all samples with $x \leq 0.13$, we find that only 10–15% of the muons experience a non-magnetic environment. These are mostly accounted for by the muons that stop outside the sample, either in the sample holder or in the walls and windows of the cryostat. The resulting doping dependence of the magnetic volume fraction is plotted in the inset of Fig. 2b. It highlights that for all samples with $x \leq 0.13$, the muons experience static magnetic fields in at least 90% of the sample volume. This does not necessarily imply that the static magnetic moments exist here in every unit cell. Nevertheless, as the stray fields from antiferromagnetic regions are known to decrease very rapidly, our data suggest that the spatial extent of such non-magnetic regions would be of the order of a few nanometres and thus of the superconducting coherence length as deduced from the upper critical field\(^\text{F12}\) and the gap magnitude\(^\text{21}\).

Next we discuss the complementary issue of the superconducting volume fractions. Transverse-field $\mu$SR measurements of the vortex state, although they establish a nearly 100% superconducting volume fraction at $x = 0.15–0.3$ (ref. 22), cannot help at $x \leq 0.13$, as the relaxation behaviour here is dominated by the magnetism that is already present at $T_c$. Nevertheless, Fig. 3a,b shows that the resistivity exhibits a fairly sharp drop towards zero and there is a sizeable diamagnetic signal in the magnetization below $T_c$. In
particular, in Fig. 3c the gap-like suppression of the infrared optical conductivity, $\sigma_{xx}$, is a clear signature of bulk superconductivity. This suppression is less pronounced at $x = 0.13$, but this may be understood due to the lower onset frequency and thus gap magnitude as marked by the arrows and the reduced condensate density. Our infrared data thus are hardly compatible with the point of view that the superconducting volume fraction is more than an order of magnitude smaller at $x = 0.13$ than at $x = 0.15$ and 0.18 where a nearly 100% superconducting volume fraction is established from $\mu$SR (ref. 22). Nevertheless, they do not enable us to accurately determine the superconducting volume fraction in these underdoped samples. Accordingly, it remains an open question whether the same or different electronic states contribute to the magnetism and to superconductivity and whether these are spatially homogeneous or phase separated on a microscopic or possibly even on a macroscopic scale, if it is limited to a very small volume fraction.

Irrespective of this open question, our data also provide evidence for a mutual interaction between these two kinds of order. This can be inferred from the doping dependence of the fitting parameters of the ZF-$\mu$SR data in Fig. 2a–c, which show that the onset of superconductivity is accompanied by pronounced changes in the magnetic state. For example, the local field, which is a measure of the magnetic order parameter, after a first drop between $x = 0$ and 0.05, remains almost constant between $x = 0.05$ and 0.1, whereas it exhibits a fairly steep decrease at $x > 0.1$ and vanishes near $x = 0.15$. The related sharp maximum in the relaxation rate around $x = 0.12=0.13$ and the decrease of the static magnetic fraction at $x > 0.10$ are furthermore indicative of a fairly sudden loss of magnetic order combined with the onset of slow dynamical fluctuations. We have already previously shown that these slow magnetic fluctuations persist to the highest achievable doping levels. Superconductivity thus seems to appear as soon as the long-range magnetic order is lost and reaches its maximal value in the presence of slow magnetic fluctuations.

Figure 4 summarizes our data in terms of a phase diagram of the superconductivity and magnetism. Also shown are recently reported data of the transition from a tetragonal to orthorhombic structure at low temperature, which track the transition temperature of the static magnetism relatively well. This observation agrees with previous reports that the structural transition is a prerequisite for the magnetic one. It shows that the nesting condition of the Fermi surface which stabilizes the SDW state depends rather sensitively on structural details, such as the Fe–As bond angle. Structural differences may also account for the remarkable differences in the phase diagram with respect to LaFeAsO$_{1-x}$F$_x$, where a recent $\mu$SR study found that magnetic correlations are limited to a much narrower doping regime and rather abruptly terminate just before superconductivity occurs at $x \geq 0.05$ (ref. 25).

A similar magnetic phase diagram has been reported in a recent neutron scattering study on CeFeAsO$_{1-x}$F$_x$ (ref. 26), where it was argued that static magnetism and superconductivity also do not coexist. Nevertheless, we note that the structural data yield a similar doping dependence of the tetragonal to orthorhombic transition as in the Sm compound. Accordingly, we expect that future studies with techniques such as $\mu$SR, which are very sensitive to weak and strongly disordered magnetism, may reveal a similar coexistence of strongly disordered magnetism and superconductivity as in SmFeAsO$_{1-x}$F$_x$.

These substantial differences in the magnetic and superconducting phase diagrams of the La and Sm compounds suggest that the magnetic correlations may have an important role in the HTSC pairing mechanism. Notably, in the Sm compound where static magnetism coexists with superconductivity and slow magnetic fluctuations persist to the highest doping levels, the maximal $T_c$ is almost double that of the La compound, where static magnetism disappears abruptly at the onset of superconductivity and spin fluctuations are hardly detectable in the superconducting samples by $\mu$SR (refs 25,27,28). Although the static magnetic correlations probably do not contribute to the superconducting pairing interaction, it seems that the slow antiferromagnetic spin fluctuations that emerge in the vicinity of an antiferromagnetic or SDW state may be very beneficial. It is indeed well established that antiferromagnetic spin fluctuations can induce or at least significantly enhance the superconducting pairing, giving rise to HTSCs with an unconventional order parameter.

In this context, we also note the similarities with the phase diagram of the cuprate HTSCs, where a comparable coexistence of magnetic correlations and superconductivity has been established. Indeed, strongly disordered static magnetism also coexists with superconductivity in the so-called strongly underdoped regime. Furthermore, in the cuprates, the onset of superconductivity gives rise to a suppression of static magnetism and the emergence of slow magnetic fluctuations that persist to higher doping levels, similar to that presented here for SmFeAsO$_{1-x}$F$_x$. For the cuprate HTSCs, these cannot be followed up to optimal doping with the $\mu$SR technique (owing to its limited time window), but they are readily seen, for example, in inelastic neutron scattering.

In summary, we have provided evidence that static magnetism in SmFeAsO$_{1-x}$F$_x$ survives to surprisingly high doping levels. Strongly disordered static magnetism and superconductivity both exist in the range of $0.1 \leq x \leq 0.13$ and prominent low-energy spin fluctuations are observed up to the highest achievable doping levels where $T_c$ is maximal. The comparison with similar structural studies suggests that the stability of the SDW state critically depends on the structural details that presumably modify the nesting condition at the Fermi surface or the frustration of competing magnetic interactions. Although many of these structural and electronic details in SmFeAsO$_{1-x}$F$_x$ are very different from those of the cuprate HTSCs, we find that the magnetic and superconducting phase diagrams as a function of doping are surprisingly similar. Our observations call for a critical examination of the widely accepted point of view that the HTSC pairing mechanism is based on strong electronic correlations in the vicinity of a Mott–Hubbard-type metal to insulator transition. The comparison with the present SmFeAsO$_{1-x}$F$_x$ superconductors rather points towards an important role of low-energy spin fluctuations that emerge on doping away from an antiferromagnetic or SDW state. A recent work has shown that the actual F content (as measured by energy dispersive X-ray analysis) of the SmFeAsO$_{1-x}$F$_x$ samples can be lower than the nominal one that is reported in our work. This would lead to a rescaling of the x axis of the phase diagram in our Fig. 4 that would not affect the conclusions presented here.

**Methods**

**Sample preparation and characterization.** Polycrystalline samples with nominal composition SmFeAsO$_{1-x}$F$_x$ with $x = 0.00, 0.05, 0.10, 0.12, 0.13, 0.15, 0.18$ and 0.30 were synthesized by conventional solid-state reaction methods as described in refs 2,11. Standard powder X-ray diffraction patterns were measured where all peaks could be indexed to the tetragonal ZrCuSiAs-type structure. d.c. resistivity and magnetization measurements were made to determine the midpoint (10–90% width) of the resistive and diamagnetic transitions with $T_c(10) = 17(8), 25(8), 38(4), 45(3)$ and 45(4) K for $x = 0.1, 0.12, 0.13, 0.15, 0.18$ and 0.3 respectively.

**Muon spin rotation.** The 100% spin-polarized muon spin rotation/relaxation ($\mu$SR) experiments were carried out using the GPS and LTF spectrometers at the Paul Scherrer Institut, Switzerland, for the low-doping samples with $0 \leq x \leq 0.15$ and using the EMU, ARGUS and MUSR spectrometers at the ISIS Facility, UK, for the samples with $0.15 \leq x \leq 0.30$. The $\mu$SR technique is especially suited for the study of magnetic and superconducting materials as it allows one to study the local magnetic field distribution on a microscopic scale and thus to directly access the volume fractions of the corresponding phases. For example, in the case of the strongly underdoped cuprate HTSCs, this technique has been very successfully applied to reveal the coexistence of weak and strongly disordered magnetism and superconductivity. Details of this technique are outlined in Supplementary Information.
References


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